Nuclear Physics

QMCPACK: Predictive and Improvable Quantum Mechanics–Based Simulations

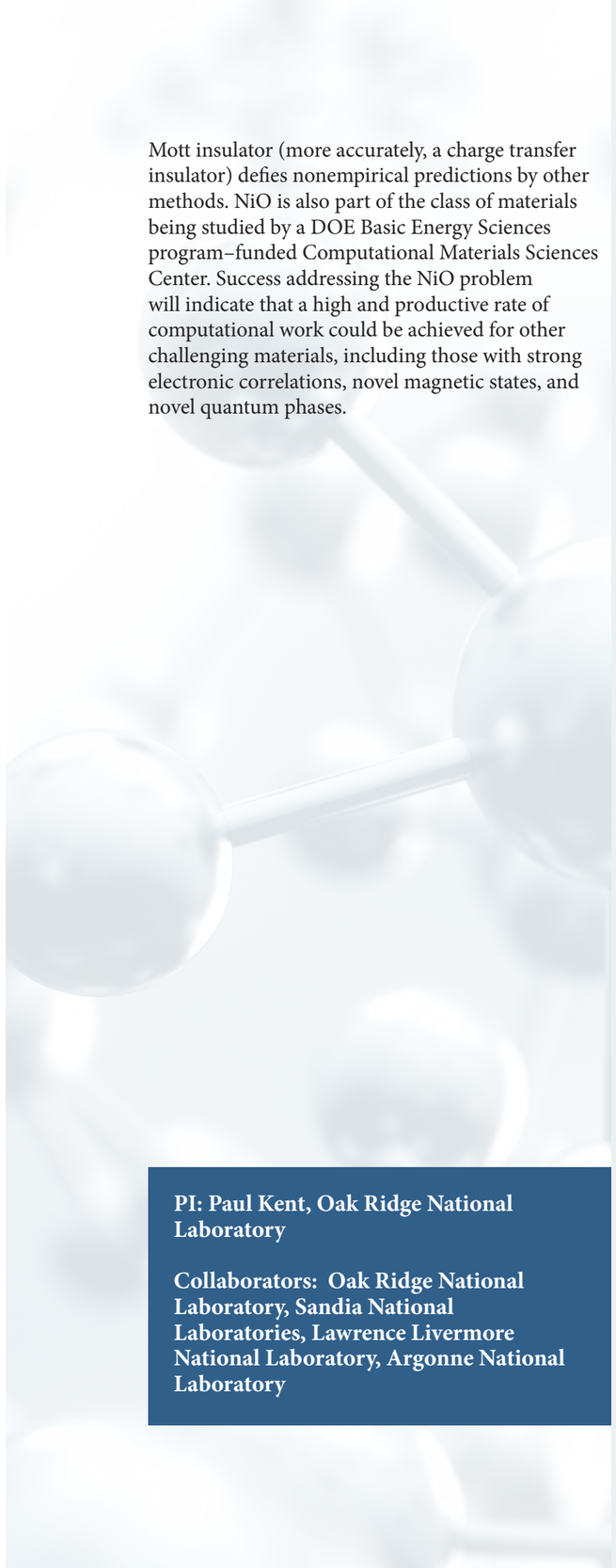
Materials design has progressed from the study of simple bulk properties to targeting collective effects in strongly correlated materials such as magnetic ordering, phase transitions, and quantum coherence. This requires a fundamentally different set of computational tools than have been used in the past. The QMCPACK project is employing QMC methods to attack this problem since they robustly deliver highly accurate calculations of complex materials that do not artificially bias solutions of a given character. Using exascale computing, QMC has the potential to attain a 18× increase in the number of modeled atoms in, as an example, metal oxide systems to fill missing gaps in experimental data and lead to advances in materials and nanoscience.

The ability to computationally design, optimize, or understand the properties of energy-relevant materials is fundamentally contingent on the existence of methods that can accurately, efficiently, and reliably simulate them. Quantum mechanics–based approaches must necessarily serve as a foundational role because only these approaches can describe matter in a truly first-principles (i.e., parameter free) and therefore robust manner. Quantum Monte Carlo (QMC) methods are ideal for the required simulations because they robustly deliver highly accurate calculations of complex materials that do not artificially bias solutions of a given character. Significantly, with increased computer power, the few approximations in these methods can be tested and systematically reduced, which is not possible with other first-principles methods.

The trade-off is that the computational demands of the QMC method are quite large. As an example, the use of petascale computers has allowed calculations of the magnetic exchange in a copper oxide, which is important for understanding the mechanism of high-temperature superconductivity. However, these calculations involved a highly symmetric supercell containing only 56 atoms, whereas a realistic model considering the defects and dopants of actual superconductors would require at least several hundreds of atoms. The 10 year challenge problem is to simulate transitional metal oxide systems of approximately 1,000 atoms to 10 meV statistical accuracy, such as complex oxide heterostructures that host novel quantum phases, using the full concurrency of exascale systems. The additional power and parallelism of exascale QMC will provide the essential predictive and quantitative capability for these and related materials that lie well beyond the capabilities of existing methods. Exascale provides the opportunity for highly impactful and enabling benchmark accuracy calculations on these materials, providing the reference calibration data that are missing from essentially all quantum mechanics–based materials calculations today. This capability will be highly useful across the materials sciences, nanoscience, and physics communities, particularly where experimental data are costly or difficult to obtain.

The challenge problem involves calculating the cohesive energy of a large supercell of nickel oxide (NiO) using QMCPACK and diffusion QMC to an accuracy of 0.010 eV per NiO formula unit at capability scale in a reasonable and scientifically productive amount of wall clock time (e.g., <1 day). The project is expected to yield a solution for a 1,024-atom supercell.

NiO has been selected because it is emblematic of science challenges addressing the complex physics of transition metal oxides. This classic Mott insulator (more accurately, a charge transfer insulator) defies nonempirical predictions by other methods. NiO is also part of the class of materials being studied by a DOE Basic Energy Sciences program–funded Computational Materials Sciences Center. Success addressing the NiO problem will indicate that a high and productive rate of computational work could be achieved for other challenging materials, including those with strong electronic correlations, novel magnetic states, and novel quantum phases.



PI: Paul Kent, Oak Ridge National Laboratory

Collaborators: Oak Ridge National Laboratory, Sandia National Laboratories, Lawrence Livermore National Laboratory, Argonne National Laboratory

Progress to date

- Developed a novel low-scaling implementation of auxiliary field QMC with reduced memory requirements and cubic scaling.
- Implemented a scheme to assemble atomic orbitals based on Gaussian basis sets that led to a 1500× reduction in memory.
- Achieved a 37× improvement on Titan using a prototype port of mainline QMCPACK developed in March 2019 to use OpenMP target/offload functionality on Summit.

QMCPACK is enabling the simulation and characterization of transition metal oxide transition systems containing up to 1,000 atoms that will improve physicists' understanding of these materials and supplement experimental investigations.